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MICROSCOPIC REGULARITIES IN THE
ELECTRICAL BREAKDOWN OF LEAD AZIDE

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ABSTRACT:

Theoretical and experimental behavior of electrical breakdown of lead azide is discussed. Data indicate it is proportional to density at lower densities, to sample thickness at higher densities and to the density-thickness product at densities over 3 g/cm^3 . Tests on samples saturated with various liquids indicate that breakdown is due to microdischarge in the pores and involves formation of electron traps. Initiation of detonation is said to be nonthermal, while its propagation is thermal, with sample density determining adequacy of heat transmission and, thus, of propagation.

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Study of electrical breakdown and excitation of detonation of primers is of interest from the point of view of explaining the elementary acts of the chemical reactions of excitation and development of rapid processes in solids [1-3]. However, a number of macroscopic factors must be taken into account in solving problems of the study of the micromechanisms of these processes, especially if the experiment is carried out with compressed polycrystalline samples. One of these processes is the formation of microdischarges in the pores of such samples [4-6], which can cause a reduction in the electrical resistance of the material and, in breakdown of the primers, cause ignition of the compound, if the size of the focus is greater than critical. Another process which reduces the electrical resistance and leads to ignition of the compound is corona discharge at the electrodes [7, 8]. An experimental criterion for revealing the presence of these macroscopic factors is direct detection of micro- and edge discharges by various means, based on recording unstable current processes [9, 10, 11]. However, even in homogeneous dielectrics and semiconductors, phenomena are observed, accompanied by appearance of high-frequency components of the current, which are connected with the discrete development of electron avalanches [12, 13]. This hinders detection of corona discharge phenomena and oscillographic duplication of the current. Another criterion for the presence of unstable discharges which change dielectric strength is the relationship of breakdown voltage to sample thickness, which can serve as a criterion for one mechanism or another [7, 8, 14].

The relationship of breakdown voltage, in the presence of microdischarges, to thickness apparently is also determined by the porosity (density) of the sample. Let us examine these patterns on the basis of the simplest model of a polycrystalline dielectric and an elementary capacitance diagram of its conversion (Fig. 1).

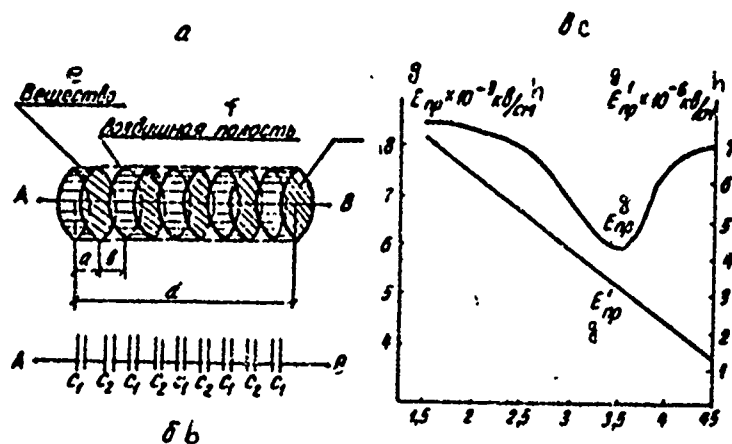


Fig. 1

Model (a), conversion scheme (b) and result of calculation of breakdown in laminar-porous dielectric.

Key: e. substance g. E_{br} , E'_{br}
f. air cavity h. kV/cm

The voltage applied to this system is distributed according to the relationship

$$U = n(E_1 a + E_2 b) \quad (1)$$

At large n (number of elements of the dielectric) the dielectric strength is determined by the first discharge in a pore, the breakdown voltage of the sample will be

$$U_{br} = n E_{br2} \left[\frac{-E_2}{\epsilon_1} a + b \right], \quad (2)$$

where E_{br2} is the dielectric strength of the air gap.

The quantities n , a and b can be expressed by the densities of the monocrystal γ_1 , air γ_2 and the sample γ , if the dimensions of the component monocrystals of the sample are known:

$$b = a \left(\frac{\gamma_1 - \gamma}{\gamma - \gamma_2} \right); \quad n = \frac{d}{a} \left(\frac{\gamma - \gamma_2}{\gamma_1 - \gamma_2} \right). \quad (3)$$

after which, expression (2) takes the form:

$$E_{br} = E_{br2} \left(\frac{\epsilon_1 - \epsilon_2}{\epsilon_1 - \epsilon_2} \right) \left[\frac{\epsilon_2}{\epsilon_1} + \left(\frac{\epsilon_1 - \epsilon_2}{\epsilon_1 - \epsilon_2} \right) \right]. \quad (4)$$

In conformance with (4), in the case of a breakdown mechanism connected with microdischarges in the pores, the dielectric strength is independent of sample thickness and is determined by its porosity and pore size (E_{br2}). The dependence of the breakdown voltage on the interelectrode distance at small thickness has been little studied at present; however, there are results in some works [6, 15], which can be extrapolated to the small thickness region.

A similar calculation for the microdischarge mechanism in a single monocrystal gives the following expression for dielectric strength

$$E'_{br} = E_{br1} \frac{\epsilon_1 - \epsilon_2}{\epsilon_1 - \epsilon_2} \left[\left(\frac{\epsilon_1 - \epsilon_2}{\epsilon_1 - \epsilon_2} \right) \frac{\epsilon_2}{\epsilon_1} + 1 \right]. \quad (5)$$

If the gas density in comparison with the density of the substance is disregarded, (4) and (5) take the following simplified forms

$$E_{br} = E_{br2} \frac{\epsilon_1}{\epsilon_1} \left[\frac{\epsilon_2}{\epsilon_1} + \frac{\epsilon_1 - \epsilon_2}{\epsilon_1} \right], \quad (6)$$

$$E'_{br} = E_{br1} \frac{\epsilon_1}{\epsilon_1} \left[\left(\frac{\epsilon_1 - \epsilon_2}{\epsilon_1} \right) \frac{\epsilon_2}{\epsilon_1} + 1 \right]. \quad (7)$$

The results of calculations by formulas (6) and (7), of the dependence of E_{br} and E'_{br} on density, with $a = 10^{-5}$ cm and $\epsilon_1 = 10$, are presented in Fig. 1. The formal calculation shows that for polycrystalline materials, there is no dependence of dielectric strength on thickness; however, there is a characteristic dependence of dielectric strength on density (the curve with the maximum for breakdown by the microdischarge mechanism in pores and the straight line for breakdown by the microdischarge mechanism in individual crystals). Still another pathway for experimental verification of the electrical breakdown macromechanism follows from the calculated model: if the breakdown mechanism is not connected with ionization in the pores, the increase in dielectric permeability of the medium ϵ_2 (for example, in dielectric saturation of the medium) should lead to a decrease in dielectric strength

of the sample and, on the other hand, if the breakdown mechanism is determined by microdischarges in the pores, increase in dielectric permeability of the medium entails increase in dielectric strength of the sample; moreover, an increase in dielectric strength should be observed as well, because of higher dielectric strength of the medium than of air.

We determined dielectric strength of the samples experimentally on the front of a standard voltage wave (1.5 μ sec/40 μ sec). The samples were tablets, compressed by a tempered steel roller in a polyvinyl chloride sheath. The contact diameter was 16 mm. The external electrode was a ball 10 mm in diameter, which was held to the outer surface of the sample in a special device. Investigation of the relationship of the breakdown voltage of the sample in this system to thickness showed that it is described well by the formula:

$$E_{br} = \frac{A}{d} + B, \quad (8)$$

which indicates the leading part of sliding discharges in breakdown in this system and the necessity for protection of the upper electrode.

Glycerin was chosen as the protective agent, as a liquid with sufficiently high conductivity, which facilitates smoothing the field, and practically does not wet the sample surface. All subsequent tests were carried out with a protected upper electrode.

The results of study of the effect of density and thickness of the disrupted layer on dielectric strength of lead azide are presented in Fig. 2, in the coordinates, breakdown voltage (V) and density (g/cm^3), at different sample thicknesses and ball electrode polarities (solid lines negative, dashed, positive polarity). The tests were carried out on at least 20 samples for each polarity. The mean arithmetic deviation is shown by the vertical marks at each experimental point. The breakdown voltage vs. density curves have a minimum at a density $\rho = 3 \text{ g/cm}^3$, with the increase in breakdown voltage more sharply expressed on the higher density side. The nature of the relationships for different polarities of the ball electrode are qualitatively similar, but the breakdown voltage with a positive potential on the ball is less. The relationships $U_{br}(h)$ and $\bar{E}_{br}(h) = U_{br}(h)/h$ were constructed from the mean values of U_{br} . It turned out that \bar{U}_{br} increases practically linearly with increase in thickness:

$$\bar{U}_{br} = A + Bh \quad (9)$$

The values of A and B, for different sample densities and ball electrode polarities are presented in the following table:

Table

$\frac{\gamma}{\rho}$	" + "		" - "	
	A	B	A	B
1,5	-	-	0,203	107,5
2	-	-	0,716	100,0
2,5	-	-	0,35	89,5
3	-0,13	106,5	-0,07	115,5
3,5	0,288	103,4	0,36	107,0
4	0,544	110,0	0,65	121,0
4,5	1,36	107,0	1,59	110,0
4,7 *	1,356	125,0	1,42	136,0

*Results of extrapolation of lead azide monocrystal density.

Key: a. γ , g/cm³

[Translator's note: commas in tabulated figures are equivalent to decimal points.]

Dielectric strength decreases with increase in thickness, for specimens with density greater than 1 g/cm³, with this relationship more clearly expressed, the higher the density (see Fig. 3).

The resulting relationships of lead azide breakdown voltage to density coincides qualitatively with the results of calculations by the approximate model of microdischarges in the pores of a complex dielectric. The dielectric strength of lead azide in the low density regions is practically independent of thickness, which also conforms to the results of the calculations. However, in proportion to approach of sample density to the density of a monocrystal, an increase in dependence of the mean breakdown gradient on thickness is observed, which does not conform to the assumptions of the predominant role of microdischarges in the pores in breakdown of polycrystalline samples. If it is considered that the cause of decrease in dielectric strength with increase in thickness is an increase in the probability of the presence of weak places in the sample, this relationship should be strengthened with increase in thickness, which also does not conform to the

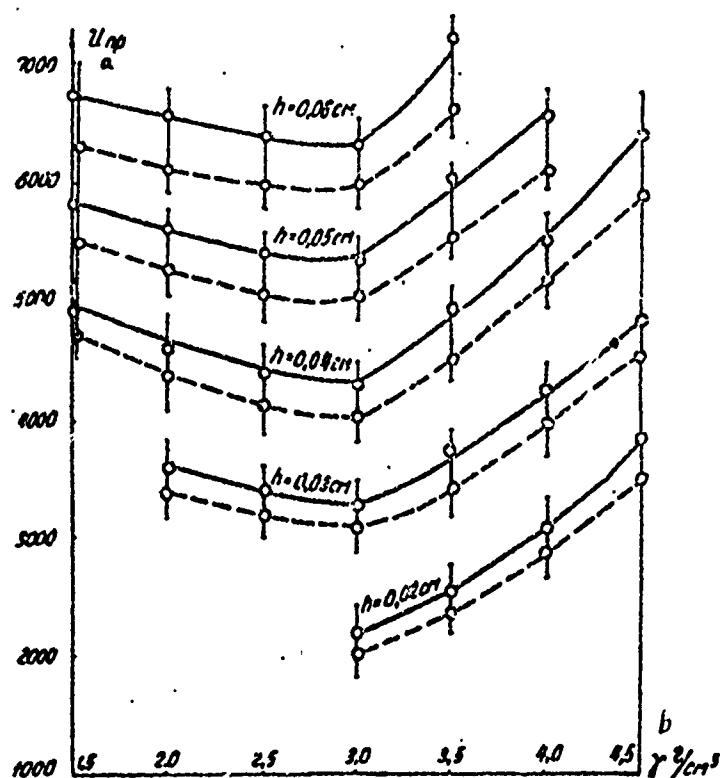


Fig. 2
Lead Azide Breakdown Voltage vs. Density and
Thickness of Disrupted Layer at Different Flat
Electrode Polarities
Key: a. U_{br} b. γ , g/cm³

results obtained. It is natural to assume that, at high density, the breakdown mechanism of the compressed polycrystalline specimen may be similar to the breakdown mechanism of solid dielectrics. By analogy with the processes of gas breakdown, the increase in density is equivalent to an increase in pressure. In both cases, this leads to a decrease in path length between impact ionization acts ϵ_1 and, consequently, to an increase in E_{br} . For purposes of ascertaining the truth of this hypothesis, U_{br} vs. the product ρh (ρh , the analog of the complex ρh in gas breakdown, is the Paschen curve [4]) was plotted. It is presented in Fig. 4. It turned out, that in the region $\rho \geq 3$ g/cm³, the breakdown voltage values are grouped in a comparatively narrow region, i.e., they are determined by product ρh , separately independent of thickness and density.

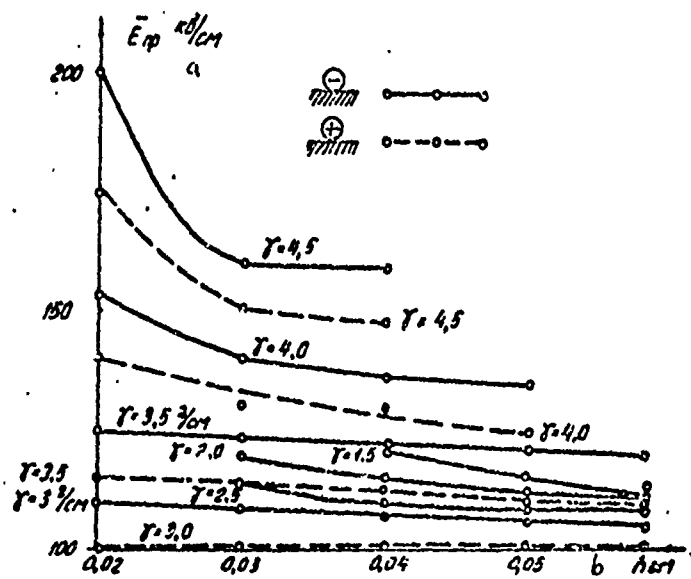


Fig. 3
Mean Breakdown Gradient of Lead Azide
vs. Density and Thickness
Key: a. \bar{E}_{br} , kV/cm b. h , cm

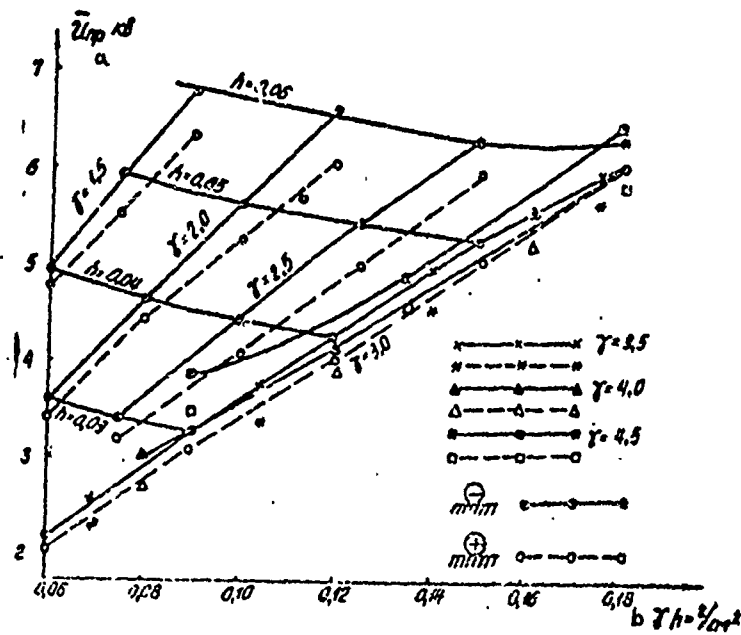


Fig. 4
Lead Azide Mean Breakdown Voltage vs.
the Combination γh
Key: a. rU_{br} , kV b. γh , g/cm²

A group of experiments also was carried out on the effect of saturation with liquids of varying dielectric properties on the breakdown voltage of compressed polycrystalline samples of lead azide. Saturation with glycerin, cresol and carbon tetrachloride does not change the nature of the function $\bar{U}_{br}(\gamma)$ very noticeably, and even the maximum breakdown voltage takes place at the same density, $\gamma = 3.0-3.5 \text{ g/cm}^3$. Saturation with transformer oil qualitatively changes the course of the curve; with decrease in density, the breakdown voltage increases monotonically. Apparently, for specimens saturated with glycerin, CCl_4 and cresol, the discharge process in the air pores is partially or completely replaced by discharge in liquid; transformer oil has a high dielectric strength, and breakdown takes place exclusively in the lead azide microcrystals. There, the calculation model satisfies the fact that dielectric strength of saturated low density components increases together with liquids, the dielectric permeability in which increases. At higher densities, its nature is the opposite. It should be noted that extrapolation of $\bar{U}_{br}(\gamma)$ curves to monocrystal densities does not reduce them to one point, which is evidence of the intervention of liquid layers in the electron stages of the breakdown.

In this manner, the results of the study permit it to be proposed that, in breakdown of samples with densities close to monocrystalline, the breakdown mechanism is connected with development of electron traps, as was proposed earlier [1] for samples of the same density $< 3 \text{ g/cm}^3$, that the process is definitely caused by microdischarges in the pores.

In study of breakdown of compressed, saturated lead azide samples, we drew attention to the fact that, in thin samples, saturated with glycerin and cresol, detonation is not excited in breakdown, and that in samples of high density, the detonation has a local nature and does not propagate to the entire charge. Experiments of Ubbelode and Woodworth [16] on the ignition of explosives in liquid of lead azide monocrystals are described in the literature. Birkomshoy and Taylor [16] consider that a high-boiling liquid decreases evolution of heat from particles and thereby suppresses thermal ignition. Thus, the phenomenon of suppression of initiation of detonation in samples of low density may have a purely thermal nature. The onset of thermal detonation in breakdown of dense saturated samples may have two causes:

1. At high densities, contact between the particles is improved and conditions for evolution of heat by this system arise;
2. Initiation of detonation during breakdown does not have a thermal nature, while its propagation is thermal.

For the purpose of determining the true cause, tests were set up on initiation of detonation in saturated samples, with a density of 4.5 g/cm^3 , by other actions, thermal and impact. In these cases, detonation also was not successfully initiated. It should be noted that saturation with glycerin is not connected with chemical reactions between glycerin and the azide: washing off the glycerin with water, with subsequent drying of the preparation, completely restores the sensitivity of lead azide to thermal pulses and shock.

The phenomena described permit the hypothesis to be made that, if electrical breakdown takes place by the microdischarge mechanism in pores or in empty defects, initiation of detonation has a purely thermal nature; in the electron trap mechanism of lead azide breakdown, the mechanism of initiation of detonation possibly does not have a thermal nature. The latter may be connected with the development of a chemical reaction between the products of shock ionization, as we pointed out earlier [1].

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